EXPERIENCE WITH *IN SITU* RADIATION MEASUREMENTS WITH THREE TYPES OF DETECTORS

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ABSTRACT

In situ radiation measurements with a plastic scintillation detector, CaF_2 scintillation detector, and a Ge γ-ray spectrometer were performed. The Monte Carlo modeling for each detector efficiency is compared with measurements for planar radioactive sources of known activity.

I. INTRODUCTION

Over the past twenty-five years, *in situ* measurements have been made of γ-ray emitting radionuclides that have contaminated the ground due to fallout, plant emissions, or facility accidents.' Commercial systems have been on the market for over ten years and are in use world wide. However, the methodology normally has been limited to the use of an unshielded NaI(T1) scintillation or a Ge semiconductor detector placed one meter above the potentially contaminated ground surface. With this classical technique the detector field-of-view extends to a radius of ≥15 m and the analysis for radioactivity concentration depends on an assumed depth profile, a uniform horizontal distribution, and a relatively flat topography. For this type of contamination distribution, an area is typically surveyed at only a limited number of measurement locations with each count being typically from 10 to 60 minutes.

When the source of the contamination is due to a small-area spill, leak, or escape of radioactivity from a disposal site, the area of ground contaminated may be initially small. With time the contamination often becomes spread both horizontally by precipitation and wind and vertically by water percolating into the ground. Such contamination is limited to a relatively small area (a few square yards to a few acres) and is usually not uniform in horizontal extent. A radiation field map made by a detector with a much smaller field-of-view provides the spatial resolution often needed to identify the individual sources of radioactivity and the direction in which the contamination plume has spread.

The present investigation reports experience gained from the use of three radiation detectors for *in situ* assay of relatively small areas of contamination. Details of the platforms used for deploying the radiation detectors and processing the contamination maps are described in other papers at this meeting.[2,3] This paper addresses the methods used to determine the detectors' efficiencies and some of the present limitations in quantitative assay.

Three detectors are used in this study. They are a) a plastic scintillator (30.5 cm x 30.5 cm x 3.8-cm thick), b) a large-area rectangular array (15.2 cm x 22.9 cm) of six CaF_2 scintillation detectors, each 7.6-cm square x 0.152-cm thick, and c) a side-shielded, thin-window, N-type Ge spectrometer (5.0 cm diameter x 2.0 cm thick) covering an energy range from ~8 keV to >1500 keV. The plastic scintillation detector is to perform gross scans for γ-ray emitting radionuclides and the CaF_2 scintillation detector is for gross scans of low energy x- and γ-ray emitting radionuclides. The Ge spectrometer is for identification and measurement of the activity concentrations of a broad range of radionuclides at specific locations.

II. DIG FACE CHARACTERIZATION SYSTEM

The purpose of our study was to develop the methodology to do qualitative and quantitative *in situ* assay for a variety of x- and γ-ray emitting radionuclides.

The results of these assays provide valuable information in near-real time for decisions concerning what ground is or remains contaminated and how the contaminated soil should be excavated and sorted for disposal during cleanup operations.

Three different platforms were used to deploy the radiation detectors. The manual push cart for covering small areas is shown in Fig. 1. A trolley, which consists of a track and bridge to provide x, and y motions, and a mast to provide motion in the vertical direction was useful for on-line assay of contaminated trenches but too limited when the plume did not remain within the width of the trolley's track. This limitation was resolved with a device that attaches to a standard excavator by replacing the shovel. It is nicknamed the "Warthog," and is shown in Fig. 2. The detectors attach to the bottom of the "Warthog" by four quick-release locking mechanisms. The "Warthog" contains the instrumentation to transmit detector position, maintain orientation of the detector even when the ground being scanned is sloped, and keeps the detector face distance to the ground nearly constant even when the ground is uneven.

III. EFFICIENCIES FOR PLANE SOURCES

The concepts involved in the calibration and data analysis processes are similar for all three detectors. The method used to provide the information needed in order to analyze the *in situ* measurements involved three steps. The first is the measurement of a detector's efficiency using point or planar radioactive sources. Plane sources up to 1.2-m square (consisting of four 0.6 m square sources[4]) could be counted with one measurement (larger source areas were simulated with multiple counts of these sources). The second is the modeling of the detector response for the measured geometries (either full-energy peak count rate or energy-loss spectrum) by means of a Monte Carlo photon and electron transport code.[5] This program, with which we have extensive experience, requires cylindrical symmetry so square detectors or sources were modeled as a circular shape preserving the area of the detector or source. Measurements are used to determine the accuracy of the model for the planar sources. Normalization to the measured values was not made in this study. Having determined the accuracy to which the detector efficiency for planar sources can be modeled, the third step involved the modeling of volume sources.

A. Plastic Scintillation Detector

The count rate of the plastic scintillation detector is recorded in a scaler for all events depositing more than ~150 keV of energy in the detector. The energy of the lower-level discriminator is determined from two separate spectral counts in which the peaks resulting from Compton scatter of the 661- and 1332-keV γ rays of ^{137}Cs and ^{60}Co, respectively, are used to deduce the energy scale. This detector scans the ground at a soil-to-detector distance of 15 to 20 cm and a scanning rate of 15 to 30 cm/s. The sides of the detector are shielded by 5.1 cm of lead, so the full field-of-view covers an area of ~60,000 cm^2 (138-cm diameter) for a ground-to-detector distance of 18.75 cm. Surveys have been made primarily over areas known to be contaminated with ^{137}Cs or ^{232}Th so our calibration efforts have focused on these nuclides. Measurements have been made for planar ^{137}Cs sources with areas of 0, 100, 3600, 14,400, and 57,600 cm^2 to verify the accuracy of the Monte Carlo based simulation/ modeling. Table 1 compares these measured efficiencies to the modeled values. As seen from the table, the agreement is better for the larger sources. This may be caused in part by the cylindrical symmetry requirement of the Monte Carlo program used.

Table 1. Comparison of measured to modeled efficiencies for plane geometries for plastic detector

	Counts per Cutoff per 100 γ's Emitted		
Source Area (cm)	Measured	Modeled	Ratio
0	2.94	2.64	1.11
3600	1.986	1.700(3)	1.17
14,400	0.821	0.759(2)	1.08
57,600	0.2327	0.2250(11)	1.03

B. Ge Semiconductor Detector

The Ge detector covers the energy range from ~8 to >1500 keV. This type detector is ideal for the detection of the L x rays emitted in the decay of many radioactinides and also detection of higher-energy γ rays. The detector is mounted in a frame which also supports a 4.44-cm thick bismuth collimator (8.89-cm inside diameter by 15-cm length) surrounding the detector housing (7.6-cm outside diameter). The typical soil-detector distance for counting specific locations is normally 23 cm (15 cm from collimator face to ground) with the detector face recessed to reduce the detector's field-of-view to ~50-cm diameter at the ground surface. Due to the time (one to 20 minutes) required to accumulate a statistically significant spectrum, this detector is not used in the scanning mode. The method of calibration and generation of information to support the analysis of *in situ* data is the same as used with the plastic scintillation detector except that radionuclide standards (e.g., ^{152}Eu) emitting γ rays over a range of energies were included in the calibration. The net areas of the full-energy spectral peaks were used in the analysis. The calibration measurements with point and planar sources and the corresponding modeling can be extensive in this case since the detector covers a range from ~8 to >1500 keV. Table 2 compares measured and modeled efficiencies. The agreement for the Ba K γ rays is not as good as for the 661 keV γ ray.

Table 2. Comparison of measured and modeled efficiencies for a plane source as a function of energy for Ge Semiconductor

Source Area (cm^2)	Photon Energy (keV)	Counts in Peak per 100 Photons Emitted: Measured	Modeled	Ratio
3,600	32–36	0.456(15)	0.603	0.76
	662	0.00619(13)	0.00686	0.90
14,400	32–36	0.0119(4)	0.0159	0.75
	662	0.00164(3)	0.00173	0.95

C. CaF_2 Detector Array

The CaF_2 detector system consists of six detectors arranged in a closely spaced 2x3 detector array. The thickness of the detector (0.152-cm thick) was chosen to give a good counting eff1ciency for the Pu L x rays and, at the same time, minimize the efficiency for higher energy (≥60 keV) K x rays and γ rays. This detector array is used in the scanning mode with a scanning rate of ≤15 cm/s for *in situ* surveys. The energies of the pulses from the L x rays and 60 keV γ rays emitted from a ^{241}Am source from each detector are aligned by adjusting the high voltage on each photomultiplier tube so that the gains are matched. This combined signal is fed into two single-channel analyzers (SCA) and counters that record the L x rays emitted by the Pu isotopes and the 59-keV γ ray emitted by ^{241}Am, respectively. The calibration of this system has, so far, been quite limited. It has been done only by measuring the counts from point sources of ^{239}Pu and ^{241}Am at various positions relative to the axis of a single detector. These results were combined to simulate the six detector array and to simulate the planar sources. Comparisons of measurements of point sources and the model calculations for the CaF_2 detector array are not yet in good agreement, with the measured values lower by ~30%. The origin of this disagreement is being explored.

IV. EFFICIENCIES FOR VOLUME SOURCES

In modeling the efficiency for volume sources a soil density of 1.5 g/cm^3 and Beck's soil composition with 10% moisture content was used.[1] The model also used only two depth distributions of contamination: a) all contamination at a given depth, and b) contamination uniformly distributed with depth.

A. Plastic Scintillation Detector

To simulate volume sources, the Monte Carlo code was used to compute the counts from disk sources of various diameters buried in soil at various depths. By averaging the results from the various depths, the expected counts from a volume source of the corresponding diameter can be calculated. These methods would allow one to simulate the count from "any" complex depth distribution for the activity, but only uniform distributions have been considered to date for reasons of practicality. The efficiency as a function of depth is shown in Table 3.

Table 3. Efficiency as a function of depth for several disk sources for the plastic detector with the discriminator set at 150 keV

Source Diameter (cm)	Depth (cm)	Counts/100 γ Emitted: Disk	Uniform with Depth to 15.2 cm
0	0.0	2.181(10)	
10	0.0	2.141	
	1.5	1.934	
	4.6	1.376	
	7.6	0.972	
	10.7	0.663	1.081
	13.7	0.462	
120	0.0	0.834	
	1.5	0.727	
	4.6	0.509	
	7.6	0.354	
	10.7	0.247	0.402
	13.7	0.173	

B. Ge Spectrometer

The efficiency as a function of depth for the Ge detector was modeled for disk sources of 10- and 120-cm diameter and is shown in Table 4.

Table 4. Efficiency as a function of depth for several disk sources for the Ge detector

		Disk Source	Volume Source, Counts/100 γ Emitted		
Source Diameter (cm)	Depth (cm)	Counts/ 100 γ Emitted	Uniform with Depth to 3 cm	Uniform with Depth to 9.2 cm	Uniform with Depth to 15.2 cm
10	0.0	0.0303			
10	1.5	0.0228	0.0228		
	4.6	0.01262		0.0142	0.00984
	7.6	0.00723			
	10.7	0.00413			
	13.7	0.00243			
120	0.0	0.00246			
120	1.5	0.00198	0.00198	0.0136	
	4.6	0.00127			0.00101
	7.6	0.000823			
	10.7	0.000596			
	13.7	0.000369			

For *in situ* measurements of ^{137}Cs, this detector can record the Ba K x rays at 32–36 keV and the 661-keV γ ray in one count. The K X/γ ratio can then be used to deduce some limits on the average depth distribution of the activity.[6]

C. CaF_2 Detector Array

Modeling calculations have been performed to investigate the expected count for ^{238}Pu contamination as a function of the depth in soil. Table 5 shows the efficiency as a function of depth in soil for one 7.6-cm square CaF_2 detector at a soil-to-detector distance of 18.18 cm. Note that the effective depth to which ^{238}Pu can be detected by the assay of the L x rays is only ~0.5 cm. Since the efficiency times mass approaches a constant for an "infinity" thick sample, this efficiency parameter can be an excellent method for measuring activity concentrations when the detected photon is an L x ray emitted from an actinide.

Table 5. Efficiency as a function of depth for several disk sources for a single CaF_2 detector

Source Radius (cm)	Source Depth (cm)	Efficiency in % for 16-keV SCA	Efficiency in % (activity uniform with depth to 0.5 cm)	Efficiency x Mass (activity uniform with depth to 0.5 cm)
10	0	1.024		
10	0.05	0.646		
10	0.15	0.245	0.206	54.6
10	0.25	0.0892		
10	0.35	0.0365		
10	0.45	0.0128		
60	0	0.155		
60	0.05	0.0716		
60	0.15	0.226	0.0209	199.2
60	0.25	0.00740		
60	0.35	0.00210		
60	0.45	0.00070		

V. FIELD MEASUREMENT EXPERIENCE

The plastic, CaF_2, and Ge detectors were used in the field at more than one site to locate, map and/or quantify ^{137}Cs, ^{232}Th, and ^{238}Pu contamination. In all cases, the radionuclide(s) causing the contamination was known. In general, this is helpful information, but it can also be acquired with the Ge spectrometer. The advantages and limitations of each platform were evaluated during this field work and the Warthog was developed for any application where an excavator was available.

The manual cart proved to be an excellent choice for use with any of the three detectors when: a) the contamination area is small and excavation has not begun, or b), specific locations of a contaminated area need to be quantified or the depth distribution estimated. Figure 3 shows a map of a small area, contaminated with ^{232}Th, that was acquired with the plastic scintillation detector mounted on the manual cart. Individual measurements are represented by dots.

The trolley proved to work well when on-line characterization of a small area such as a canal or trench-shaped area is being remediated. The trolley is limited in that its track must be laid on a horizontal surface besides the trench prior to excavation. When the breath or depth of the contaminated area extends beyond the trolley's reach, it cannot be easily reoriented.

The Warthog platform was developed because it could easily follow the contamination no matter its distribution. The Warthog attaches to the arm of an excavator in place of the shovel. It has the capability to

Maintain a detector at a predetermined height above the ground surface and communicate its position and detector count rate(s) every second via radio frequency transmission. The operator of the excavator is equipped with a lap top computer that displays the detector's location and path as a swath on the monitor screen. From the locations and count rates a matrix of measurements are converted into a contour map.

VI. CONCLUSIONS

The three different types of detector systems discussed here have proved very useful in a variety of *in situ* measurement situations. It is clear that the continuous survey capabilities of the plastic scintillator and CaF_2 detectors are of great value since they reduce the chance of missing small "hot spots" of contamination, a limitation of any method involving sampling. The use of side-shielded detectors for the scanning mode provides better control of the detector's field-of-view and better spatial resolution.

The use of Monte Carlo modeling/simulation as a means of computing the expected count rates from different potential source distributions allows one to compare different potential contamination distributions in extent and depth and thereby make realistic estimates of the range of contamination levels possible. We believe that the need to remediate radioactively contaminated areas in a cost effective manner will encourage further development of these *in situ* characterization techniques.

The results of the above efficiency determinations are encouraging but limitations exist. Modeling of the scintillation detector efficiencies did not incorporate the influence of the light collection and photomultiplier response on the observed spectrum which may partially explain the large difference between the measured and modeled efficiencies for the CaF_2 detector.

The dimensions of the Ge crystal, and the material composition and dimensions of the structural hardware are not always known and often are proprietary. This impacts the accuracy attainable from modeling the Ge detector efficiency. In the present study the Ge detector was an N-type with a thin carbon-fiber window with the sides shielded by the collimator. Therefore, these effects were considered small.

Although the absorption cross sections need not be known precisely at γ-ray energies above a few hundred keV for soil matrixes, the absorption cross sections, the soil composition, densities, and moisture content must be known precisely for accurate modeling of the low-energy L x rays. This may require obtaining soil composition, density and moisture measurements at the remediation site. In spite of these limitations on the accuracy of low-energy measurements, the CaF_2 detector array can readily identify "hot spots" of actinide contamination that might be missed by a sampling protocol used to release a remediated site for other use. The lower-level-of-detection, LLD, for ^{238}Pu achievable with this six-detector array has been deduced from a comparison of *in situ* measurements with soil samples analyzed in a laboratory. For a scanning mode of 15 cm/s with multiple counts covering the detector field of view, the critical level of detection is $L_c \sim 126$ pCi/g and the lower-limit-of-detection (LLD) is ~ 290 pCi/g for activity evenly distributed within at least the top 0.5 cm of the soil surfaced

The background count rate for all detectors was assumed to be constant during any set of gross count-rate measurements which is not typical in actual practice. This assumption introduces only a small error when the count rates from the contamination are a factor of ≥ 5 than the background count rate.

Finally, the largest impact on accuracy- is the distribution of the activity. The depth distribution of the contamination is usually the most difficult to determine; however, a change in the extent of the horizontal distribution within the detector's field-of-view may be more serious.

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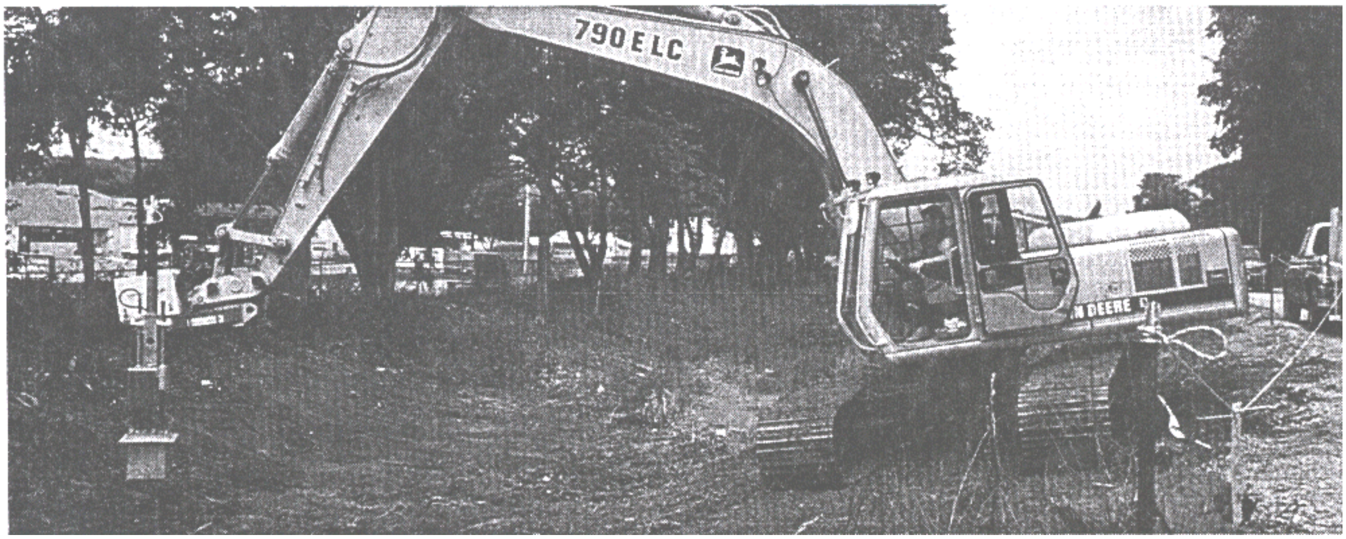
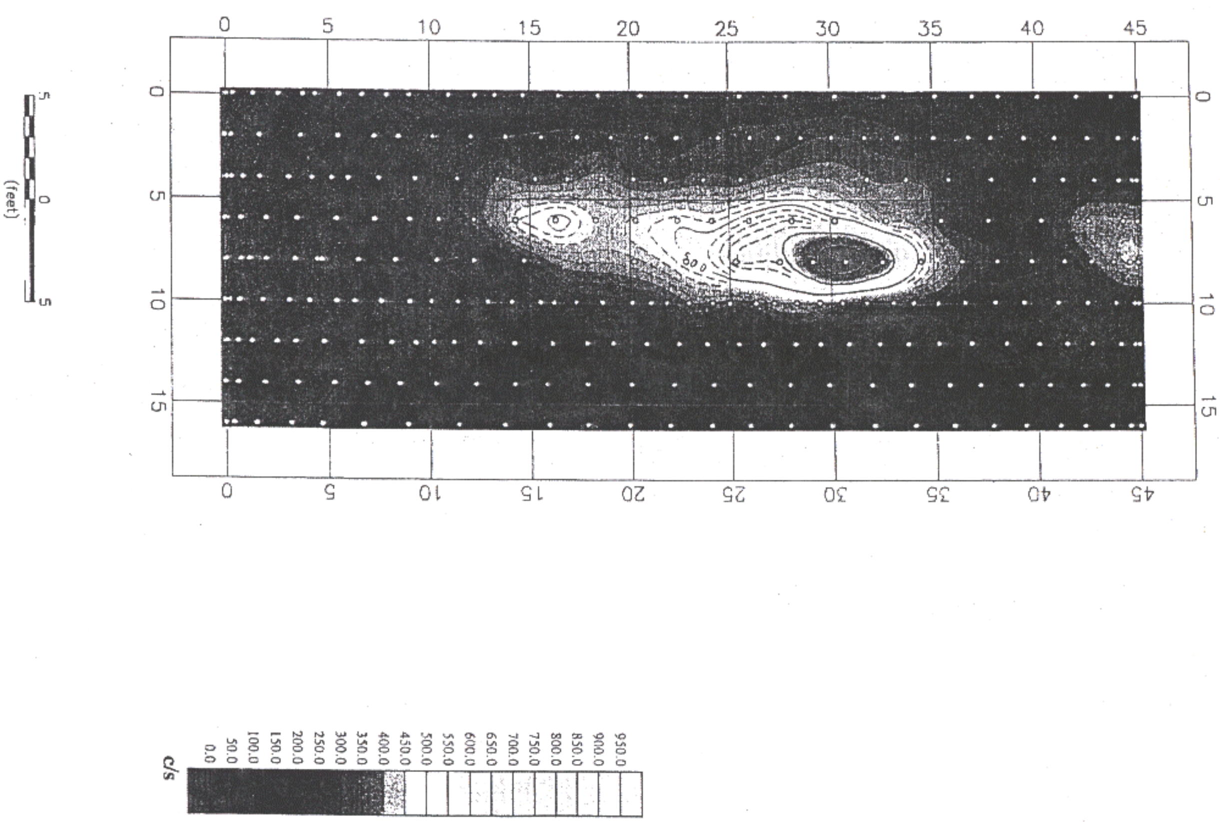
1. Manual push cart.



2. Warthog in operation.



3. Map of a small area of ground contaminated with ^{232}Th.